



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/GB00/01646 <b>(22) International Filing Date:</b> 28 April 2000 (28.04.00) <b>(30) Priority Data:</b> 9909956.6                      29 April 1999 (29.04.99)                      GB <b>(71) Applicant (for all designated States except US):</b> ASTON UNIVERSITY [GB/GB]; Aston Triangle, Birmingham B4 7ET (GB). <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only):</b> AL-MALAIKA, Sahar [GB/GB]; 55 Monmouth Drive, Sutton Coldfield B73 6JH (GB). <b>(74) Agent:</b> EYLES, Christopher, Thomas; W.P. Thompson & Co., Celcon House, 289-293 High Holborn, London WC1V 7HU (GB).		<b>(81) Designated States:</b> AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the</i> <i>claims and to be republished in the event of the receipt of</i> <i>amendments.</i>
<b>(54) Title:</b> THERMOPLASTIC MOULDING COMPOSITIONS AND POLYMER ADDITIVES		
<b>(57) Abstract</b> <p>A polymer additive is described for addition to a thermoplastic moulding composition comprising polyethylene terephthalate or a copolyester thereof so as to effect reduction of the level of acetaldehyde resulting after processing thereof, said polymer additive comprising a hydroxylic compound selected from aliphatic hydroxylic compounds containing at least two hydroxy groups, aliphatic-cycloaliphatic compounds containing at least two hydroxy groups, and cycloaliphatic hydroxylic compounds containing at least two hydroxy groups, uniformly distributed in a polyester-compatible organic liquid carrier. The invention further relates to the use of a hydroxylic compound selected from aliphatic hydroxylic compounds containing at least two hydroxy groups, aliphatic-cycloaliphatic compounds containing at least two hydroxy groups, and cycloaliphatic hydroxylic compounds containing at least two hydroxy groups, as an additive to a thermoplastic moulding composition for the reduction of the amount of acetaldehyde formed upon subjecting said moulding composition to melt processing. Thermoplastic moulding compositions, processes using same and preforms and bottles made therefrom are also described. Preferred hydroxylic compounds include triglycerin, trimethylolpropane, dipentaerythritol, tripentaerythritol, D-mannitol, D-sorbitol, and xylitol.</p>		

CLAIMS:

1. Use of a hydroxylic compound selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic compounds and cycloaliphatic compounds containing at least two hydroxy groups for reducing the level of acetaldehyde resulting after processing of a thermoplastic moulding composition comprising polyethylene terephthalate or a copolyester thereof.
2. Use according to claim 1, in which the hydroxylic compound contains from 3 to about 8 hydroxy groups.
3. Use according to claim 1 or claim 2 in which in the hydroxylic compound contains a pair of hydroxy groups attached to respective carbon atoms which are separated one from another by a single carbon atom.
4. Use according to anyone of claims 1 to 3, in which said hydroxylic compound is selected from triglycerin, trimethylolpropane, dipentaerythritol, tripentaerythritol, D-mannitol, D-sorbitol, and xylitol.
5. Use according to any one of claims 1 to 4, in which the hydroxylic compound is manufactured as a polymer additive for addition to the thermoplastic moulding composition.
6. Use according to claim 5, in which said polymer additive comprises said hydroxylic compound uniformly distributed in a liquid carrier.
7. Use according to claim 6, in which the hydroxylic

compound:liquid carrier weight ratio ranges from about 0.1:1 to about 1.5:1.

8. Use according to claim 6 or claim 7, in which the liquid carrier is a polyester-compatible organic oil-based vehicle.

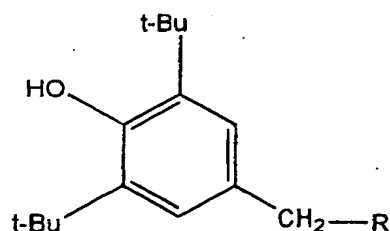
9. Use according to any one of claims 5 to 8, in which the polymer additive further comprises at least one polyester-compatible colorant.

10. Use according to any one of claims 5 to 9, in which the polymer additive further comprises an antioxidant.

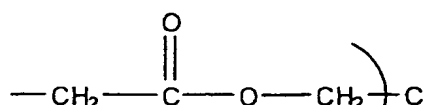
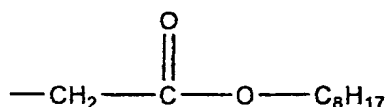
11. Use according to claim 10, in which the antioxidant is a hindered phenol antioxidant.

12. Use according to claim 11, in which the antioxidant is a 4-substituted-2,6-di-tertiary butyl phenol or an  $\alpha$ -tocopherol.

13. Use according to claim 12, in which the antioxidant has the formula:

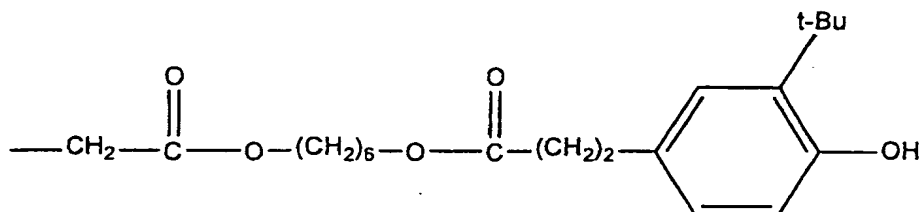


in which R is hydrogen,

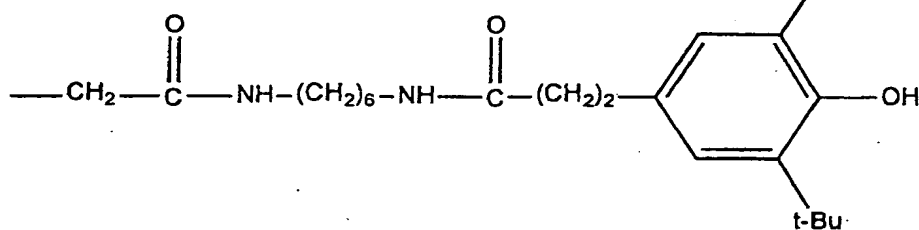


Amended claims

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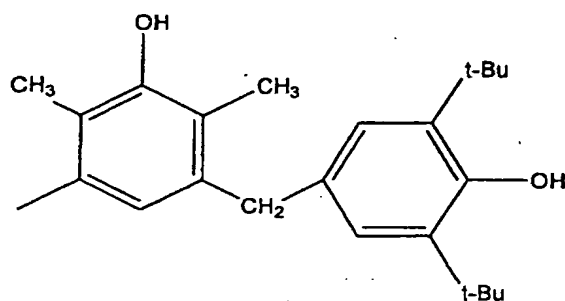


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or

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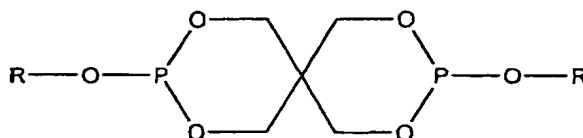
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14. Use according to claim 12, in which the antioxidant comprises synthetic Vitamin E.

25 15. Use according to any one of claims 5 to 14, in which the polymer additive further comprises a phosphite antioxidant.

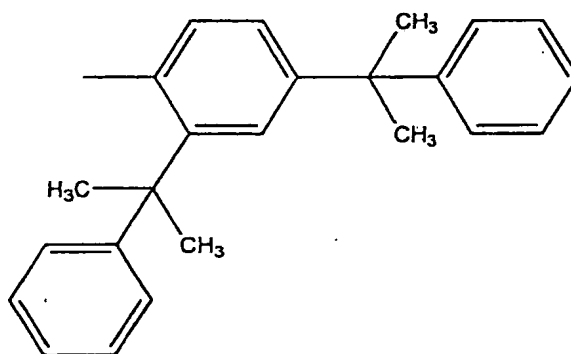
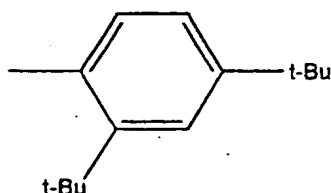
16. Use according to claim 15, in which the phosphite antioxidant has the structure:

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in which R is



or  $C_{18}H_{37}$ .

17. Use according to any one of claims 1 to 16, in which the processing of the thermoplastic moulding composition results in the formation of bottles or bottle preforms.

18. A thermoplastic moulding composition comprising a polymer component comprising polyethylene terephthalate or a copolyester thereof and an amount effective to cause reduction of the level of acetaldehyde resulting after processing thereof of a hydroxylic compound uniformly distributed therein, said hydroxylic compound containing from 3 to about 8 hydroxy groups and being selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic compounds, and cycloaliphatic hydroxylic compounds and the amount of said hydroxylic compound ranging from about 0.0001 % by weight up to about 2 % by weight based upon

the weight of the polymer component.

19. A thermoplastic moulding composition according to claim 18, in which the hydroxylic compound contains a pair  
5 of hydroxy groups attached to respective carbon atoms which are separated one from another by a single carbon atom.

20. A thermoplastic moulding composition according to claim 18 or claim 19, in which said hydroxylic compound is  
10 selected from triglycerin, trimethylolpropane, dipentaerythritol, and tripentaerythritol.

21. A thermoplastic moulding composition according to any one of claims 18 to 20, further comprising an  
15 antioxidant.

22. A thermoplastic moulding composition according to claim 21, in which the amount of antioxidant compound  
20 ranges from about 0.0001 % by weight up to about 2 % by weight based upon the weight of the polymer component.

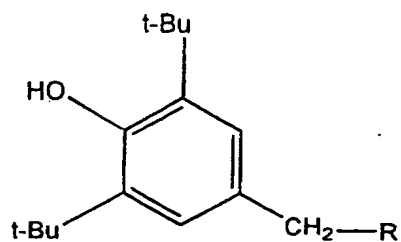
23. A thermoplastic moulding composition according to claim 21 or claim 22, in which the antioxidant is a  
25 hindered phenol antioxidant.

24. A thermoplastic moulding composition according to claim 23, in which the antioxidant is a 4-substituted-2,6-di-tertiary butyl phenol or an  $\alpha$ -tocopherol.  
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25. A thermoplastic moulding composition according to claim 24, in which the antioxidant has the formula:

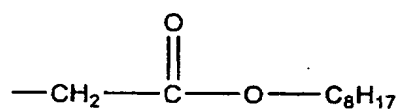
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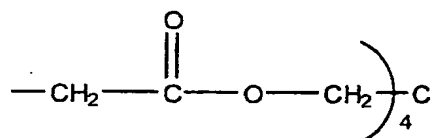


in which R is hydrogen,

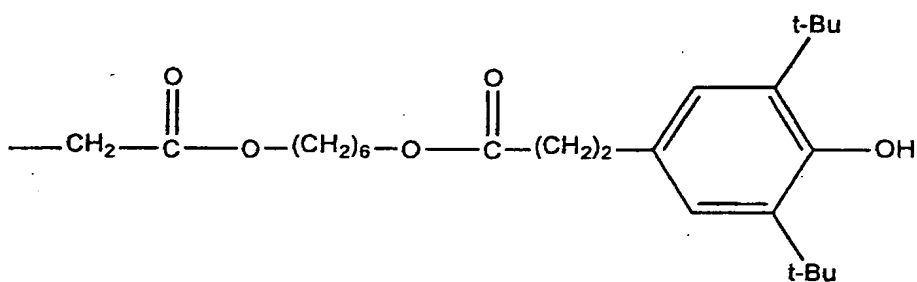
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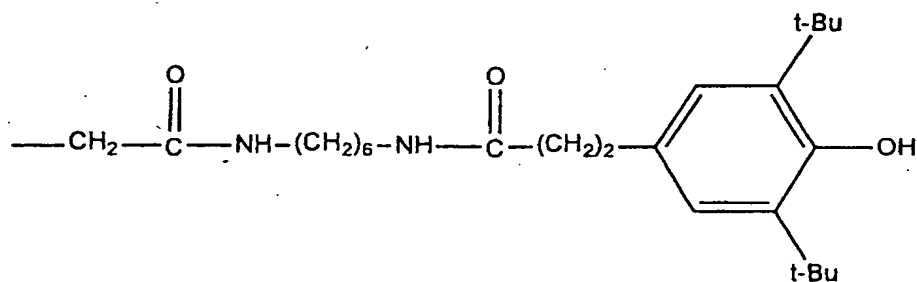
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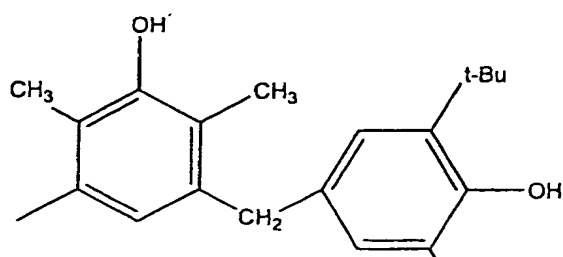


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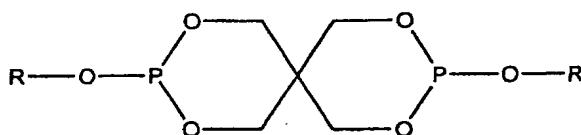


Amended claims

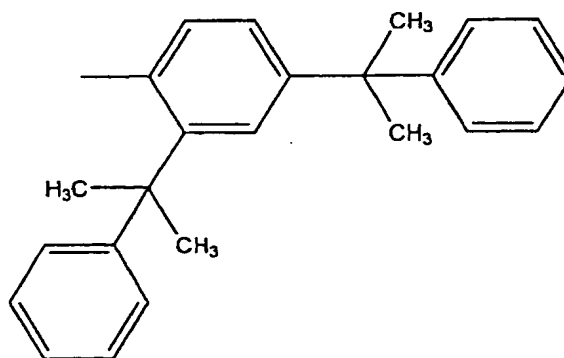
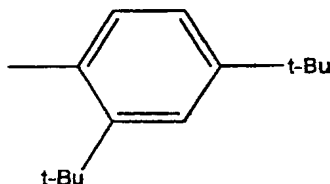
26. A thermoplastic moulding composition according to claim 24, in which the antioxidant comprises synthetic Vitamin E.

27. A thermoplastic moulding composition according to any one of claims 21 to 26, in which the antioxidant comprises a phosphite antioxidant.

28. A thermoplastic moulding composition according to claim 27, in which the phosphite antioxidant has the structure:



in which R is



or  $C_{18}H_{37}$ .



29. A thermoplastic moulding composition according to any one of claims 18 to 28, further comprising a minor amount of a liquid carrier for said hydroxylic compound.

5 30. A process for the production of a moulded article which comprises providing a thermoplastic moulding composition comprising polyethylene terephthalate or a copolyester thereof having an amount effective to cause  
10 reduction of the level of acetaldehyde resulting after processing thereof of a hydroxylic compound uniformly distributed therein, said hydroxylic compound containing from 3 to about 8 hydroxy groups and being selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic hydroxylic compounds, and cycloaliphatic hydroxylic  
15 compounds, and the amount of said hydroxylic compound ranging from about 0.0001 % by weight up to about 2 % by weight based upon the weight of the polymer component, and subjecting said thermoplastic moulding composition to an injection moulding step thereby to form a moulded  
20 article.

31. A process according to claim 30, in which the moulded article is a preform for use in a subsequent blow  
25 moulding step to form a bottle.

32. A process according to claim 30 or claim 31, in which the hydroxylic compound contains a pair of hydroxy groups attached to respective carbon atoms which are  
30 separated one from another by a single carbon atom.

33. A process according to any one of claims 30 to 32, in which said hydroxylic compound is selected from triglycerin, trimethylolpropane, dipentaerythritol, and

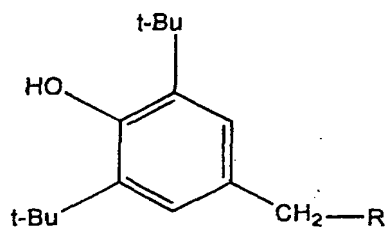
tripentaerythritol.

34. A process according to any one of claims 30 to 33,  
in which the thermoplastic moulding composition further  
5 comprises an antioxidant.

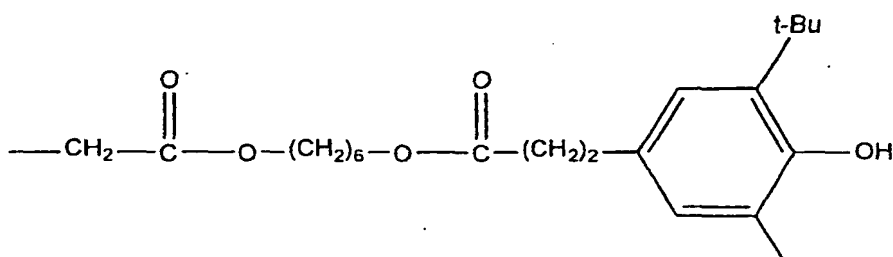
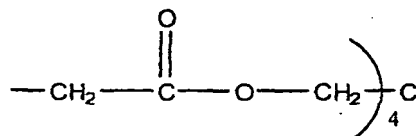
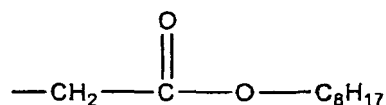
35. A process according to claim 34, in which the  
antioxidant is a hindered phenol antioxidant.

10 36. A process according to claim 35, in which the  
antioxidant is a 4-substituted-2,6-di-tertiary butyl  
phenol or an  $\alpha$ -tocopherol.

15 37. A process according to claim 36, in which the  
antioxidant has the formula:

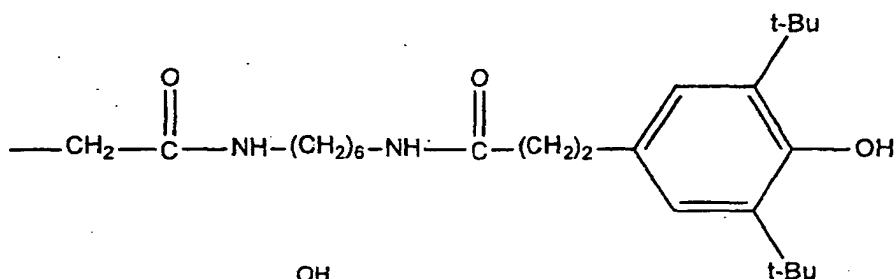


in which R is hydrogen,

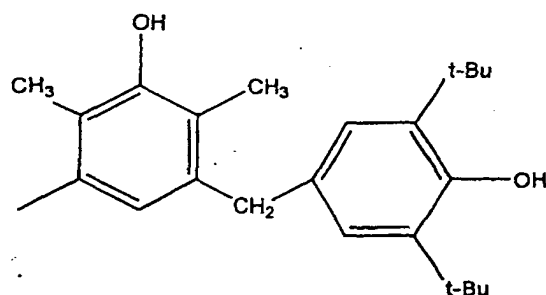


Amended claims

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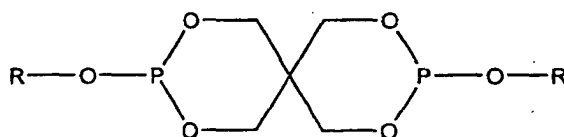
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38. A process according to claim 36, in which the antioxidant comprises synthetic Vitamin E.

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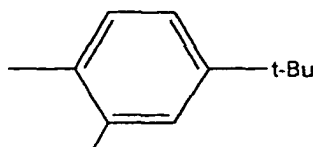
39. A process according to any one of claims 34 to 38, in which the antioxidant comprises a phosphite antioxidant.

25 40. A process according to claim 39, in which the phosphite antioxidant has the structure:



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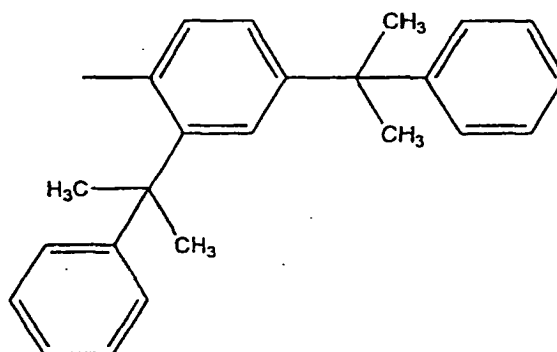
in which R is



Amended claims

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or  $C_{18}H_{37}$ .

41. A process according to any one of claims 30 to 40,  
15 in which the thermoplastic moulding composition further comprises at least one polyester-compatible colorant.

42. A process according to any one of claims 30 to 41,  
20 in which the thermoplastic moulding composition further comprises a minor amount of a liquid carrier for said aliphatic hydroxylic compound.

43. Moulded articles made by a process according to any  
25 one of claims 30 to 42.

44. Moulded articles according to claim 43, which are  
preforms for bottles.

45. Bottles blow moulded from a preform according to  
30 claim 44.

46. An injection moulded bottle preform comprising a  
polymer component comprising polyethylene terephthalate or

a copolyester thereof and from about 0.0001% by weight up to about 2% by weight based upon the weight of the polymer component of a hydroxylic compound uniformly distributed therein, said hydroxylic compound containing from 3 to  
5 about 8 hydroxy groups and being selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic compounds and cycloaliphatic compounds.

47. An injection moulded bottle preform according to  
10 claim 46, in which the amount of acetaldehyde present in the preform is from 29 to 70% less than that in a comparison preform prepared under identical conditions but with the omission of the hydroxylic compound.

15 48. An injection moulded bottle preform according to claim 46 or claim 47, in which there is incorporated a polyester-compatible colorant.

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CLAIMS:

1. Use of a hydroxylic compound selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic compounds  
5 and cycloaliphatic compounds containing at least two hydroxy groups for reducing the level of acetaldehyde resulting after processing of a thermoplastic moulding composition comprising polyethylene terephthalate or a copolyester thereof.
- 10 2. Use according to claim 1, in which the hydroxylic compound contains from 3 to about 8 hydroxy groups.
- 15 3. Use according to claim 1 or claim 2 in which in the hydroxylic compound contains a pair of hydroxy groups attached to respective carbon atoms which are separated one from another by a single carbon atom.
- 20 4. Use according to anyone of claims 1 to 3, in which said hydroxylic compound is selected from triglycerin, trimethylolpropane, dipentaerythritol, tripentaerythritol, D-mannitol, D-sorbitol, and xylitol.
- 25 5. Use according to any one of claims 1 to 4, in which the hydroxylic compound is manufactured as a polymer additive for addition to the thermoplastic moulding composition.
- 30 6. Use according to claim 5, in which said polymer additive comprises said hydroxylic compound uniformly distributed in a liquid carrier.
7. Use according to claim 6, in which the hydroxylic

compound:liquid carrier weight ratio ranges from about 0.1:1 to about 1.5:1.

8. Use according to claim 6 or claim 7, in which the liquid carrier is a polyester-compatible organic oil-based vehicle.

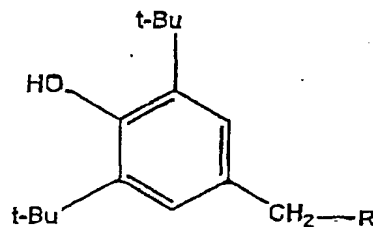
9. Use according to any one of claims 5 to 8, in which the polymer additive further comprises at least one polyester-compatible colorant.

10. Use according to any one of claims 5 to 9, in which the polymer additive further comprises an antioxidant.

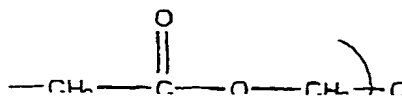
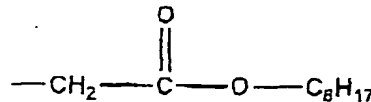
11. Use according to claim 10, in which the antioxidant is a hindered phenol antioxidant.

12. Use according to claim 11, in which the antioxidant is a 4-substituted-2,6-di-tertiary butyl phenol or an  $\alpha$ -tocopherol.

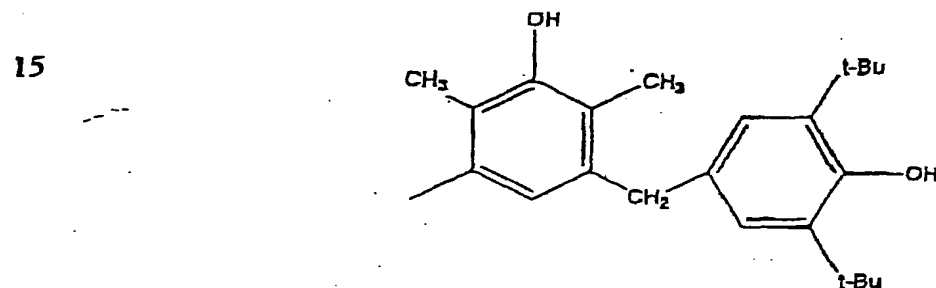
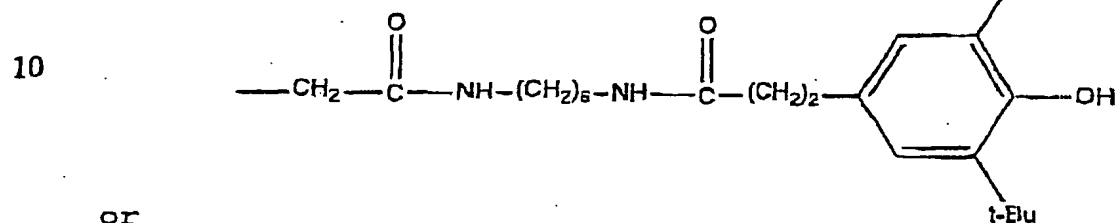
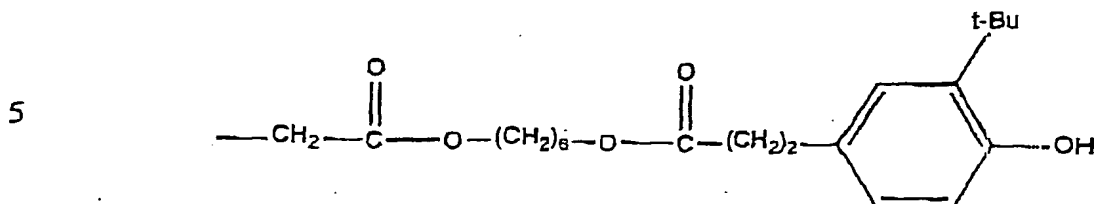
13. Use according to claim 12, in which the antioxidant has the formula:



in which R is hydrogen,



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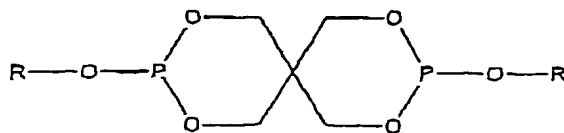


14. Use according to claim 12, in which the antioxidant comprises synthetic Vitamin E.

25 15. Use according to any one of claims 5 to 14, in which the polymer additive further comprises a phosphite antioxidant.

16. Use according to claim 15, in which the phosphite antioxidant has the structure:

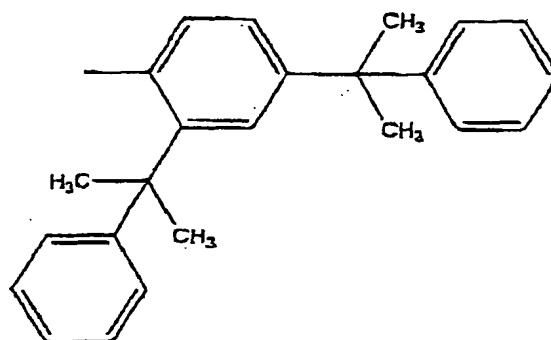
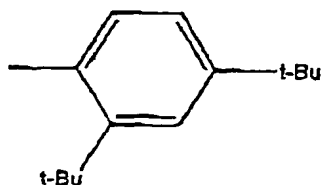
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in which R is



or  $C_{18}H_{37}$ .

17. Use according to any one of claims 1 to 16, in which the processing of the thermoplastic moulding composition results in the formation of bottles or bottle preforms.

18. A thermoplastic moulding composition comprising a polymer component comprising polyethylene terephthalate or a copolyester thereof and an amount effective to cause reduction of the level of acetaldehyde, resulting after processing thereof of a hydroxylic compound uniformly distributed therein, said hydroxylic compound containing from 3 to about 8 hydroxy groups and being selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic compounds, and cycloaliphatic hydroxylic compounds and the amount of said hydroxylic compound ranging from about 0.0001 % by weight up to about 2 % by weight based upon

the weight of the polymer component.

19. A thermoplastic moulding composition according to claim 18, in which the hydroxylic compound contains a pair of hydroxy groups attached to respective carbon atoms which are separated one from another by a single carbon atom.

20. A thermoplastic moulding composition according to claim 18 or claim 19, in which said hydroxylic compound is selected from triglycerin, trimethylolpropane, dipentaerythritol, and tripentaerythritol.

21. A thermoplastic moulding composition according to any one of claims 18 to 20, further comprising an antioxidant.

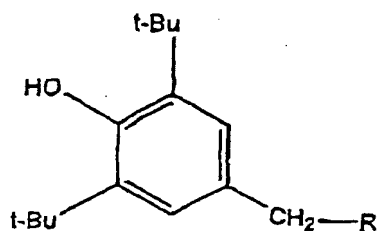
22. A thermoplastic moulding composition according to claim 21, in which the amount of antioxidant compound ranges from about 0.0001 % by weight up to about 2 % by weight based upon the weight of the polymer component.

23. A thermoplastic moulding composition according to claim 21 or claim 22, in which the antioxidant is a hindered phenol antioxidant.

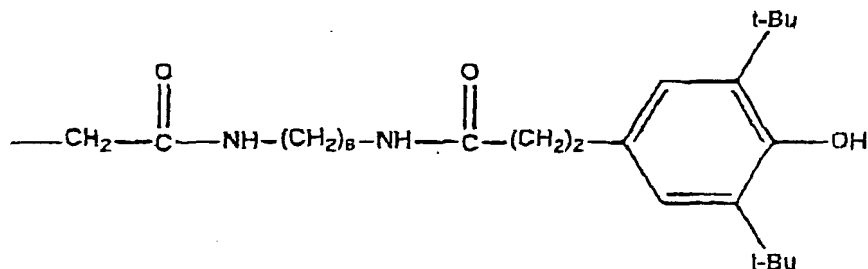
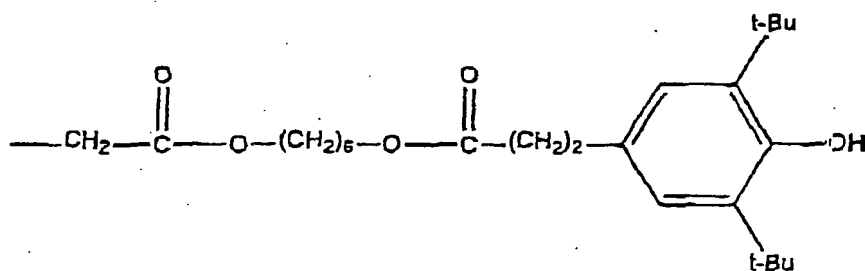
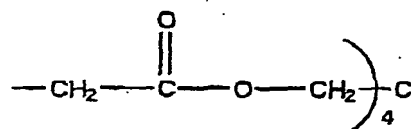
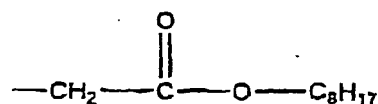
24. A thermoplastic moulding composition according to claim 23, in which the antioxidant is a 4-substituted-2,6-di-tertiary butyl phenol or an  $\alpha$ -tocopherol.

25. A thermoplastic moulding composition according to claim 24, in which the antioxidant has the formula:

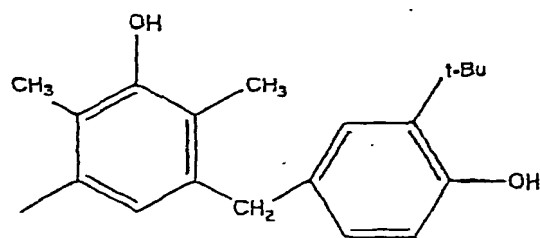
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in which R is hydrogen,



or



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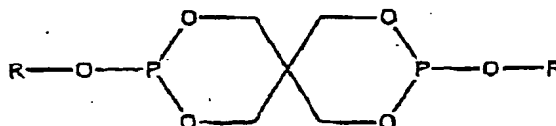
26. A thermoplastic moulding composition according to claim 24, in which the antioxidant comprises synthetic Vitamin E.

27. A thermoplastic moulding composition according to any one of claims 21 to 26, in which the antioxidant comprises a phosphite antioxidant.

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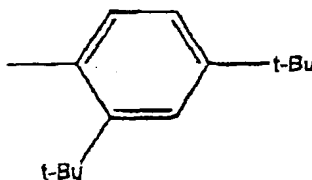
28. A thermoplastic moulding composition according to claim 27, in which the phosphite antioxidant has the structure:

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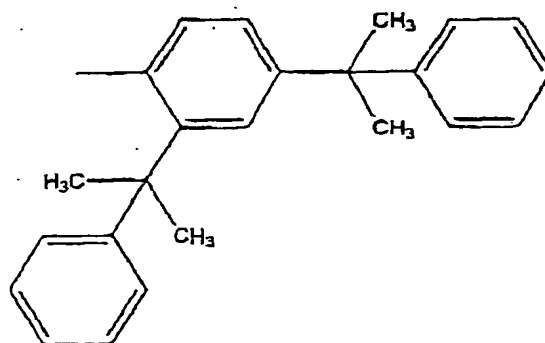


in which R is

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or  $C_{18}H_{37}$ .

29. A thermoplastic moulding composition according to any one of claims 18 to 28, further comprising a minor amount of a liquid carrier for said hydroxylic compound.

5 30. A process for the production of a moulded article which comprises providing a thermoplastic moulding composition comprising polyethylene terephthalate or a copolyester thereof having an amount effective to cause  
10 reduction of the level of acetaldehyde resulting after processing thereof of a hydroxylic compound uniformly distributed therein, said hydroxylic compound containing from 3 to about 8 hydroxy groups and being selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic hydroxylic compounds, and cycloaliphatic hydroxylic  
15 compounds, and the amount of said hydroxylic compound ranging from about 0.0001 % by weight up to about 2 % by weight based upon the weight of the polymer component, and subjecting said thermoplastic moulding composition to an injection moulding step thereby to form a moulded  
20 article.

31. A process according to claim 30, in which the moulded article is a preform for use in a subsequent blow moulding step to form a bottle.

25

32. A process according to claim 30 or claim 31, in which the hydroxylic compound contains a pair of hydroxy groups attached to respective carbon atoms which are separated one from another by a single carbon atom.

30

33. A process according to any one of claims 30 to 32, in which said hydroxylic compound is selected from triglycerin, trimethylolpropane, dipentaerythritol, and

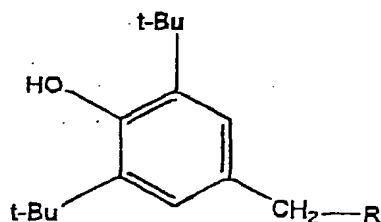
tripentaerythritol.

34. A process according to any one of claims 30 to 33,  
in which the thermoplastic moulding composition further  
5 comprises an antioxidant.

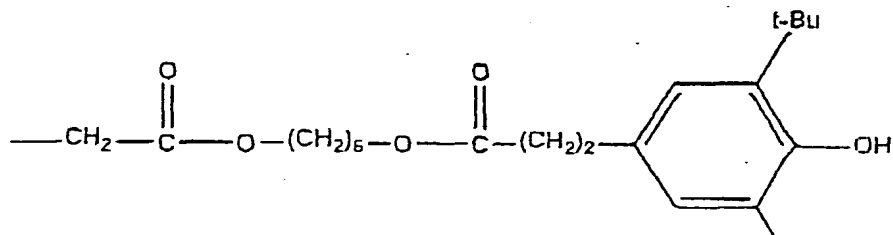
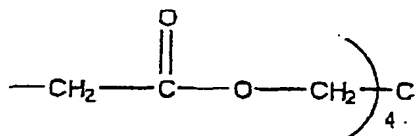
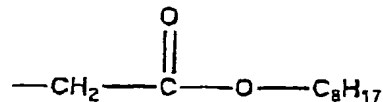
35. A process according to claim 34, in which the  
antioxidant is a hindered phenol antioxidant.

10 36. A process according to claim 35, in which the  
antioxidant is a 4-substituted-2,6-di-tertiary butyl  
phenol or an  $\alpha$ -tocopherol.

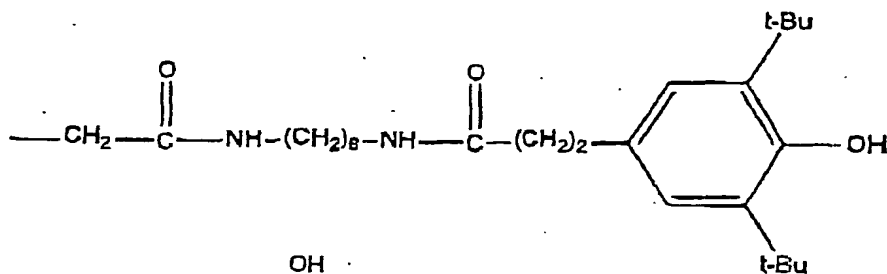
15 37. A process according to claim 36, in which the  
antioxidant has the formula:



in which R is hydrogen,

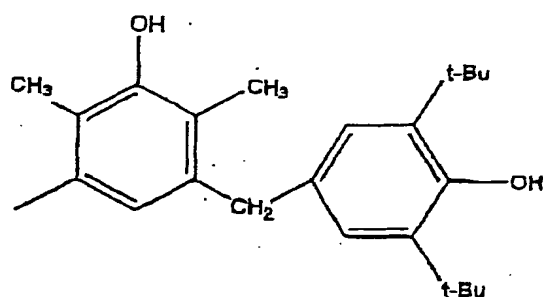


5



10 or

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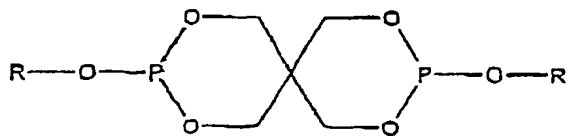
38. A process according to claim 36, in which the antioxidant comprises synthetic Vitamin E.

20

39. A process according to any one of claims 34 to 38, in which the antioxidant comprises a phosphite antioxidant.

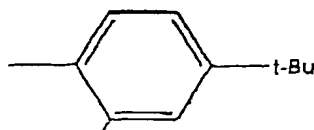
25

40. A process according to claim 39, in which the phosphite antioxidant has the structure:

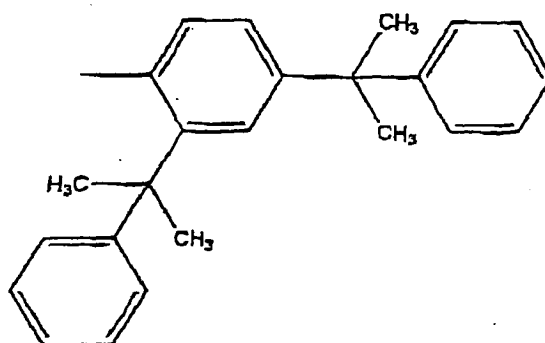


30

in which R is



43



or C<sub>18</sub>H<sub>17</sub>.

41. A process according to any one of claims 30 to 40,  
in which the thermoplastic moulding composition further  
comprises at least one polyester-compatible colorant.

42. A process according to any one of claims 30 to 41,  
in which the thermoplastic moulding composition further  
comprises a minor amount of a liquid carrier for said  
aliphatic hydroxylic compound.

43. Moulded articles made by a process according to any  
one of claims 30 to 42.

44. Moulded articles according to claim 43, which are  
preforms for bottles.

45. Bottles blow moulded from a preform according to  
claim 44.

46. An injection moulded bottle preform comprising a  
polymer component comprising polyethylene terephthalate or



a copolyester thereof and from about 0.0001% by weight up to about 2% by weight based upon the weight of the polymer component of a hydroxylic compound uniformly distributed therein, said hydroxylic compound containing from 3 to  
5 about 8 hydroxy groups and being selected from aliphatic hydroxylic compounds, aliphatic-cycloaliphatic compounds and cycloaliphatic compounds.

47. An injection moulded bottle preform according to  
10 claim 46, in which the amount of acetaldehyde present in the preform is from 29 to 70% less than that in a comparison preform prepared under identical conditions but with the omission of the hydroxylic compound.

15 48. An injection moulded bottle preform according to claim 46 or claim 47, in which there is incorporated a polyester-compatible colorant.

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